



Multi-scale Modeling of Structural Modification in Fuel

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Outline

1. Cradle-to-Grave Fuel Microstructure

- *Fresh fuel characteristics*
- *Microstructure evolution in reactor (swelling, gas kinetics, rim structure)*

2. Existing Fuel Performance Models

- *Thermal conductivity*
- *Fission gas release*
- *Volume change*

3. Modeling and Simulation

- *Review of techniques*
- *Mesoscale modeling of gas bubble evolution & thermal conductivity*
- *Scale-bridging to macroscale (whole pellet)*

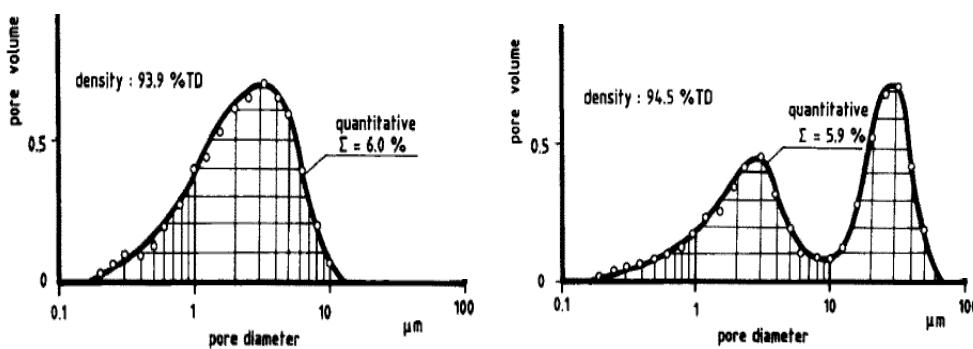
Fresh Fuel



Fabrication: powder-compact, sintering

Dimensions: diameter (~ 1 cm), height (~ 2 cm)

Enrichment: ~ 5 % (U^{235})



Grain Size: ~ 10 μm

Pore Size: ~ 3 μm (uni-modal)

~ 3 μm & 20 μm (bi-modal)

Density: ~ 95%

Fuel Microstructure

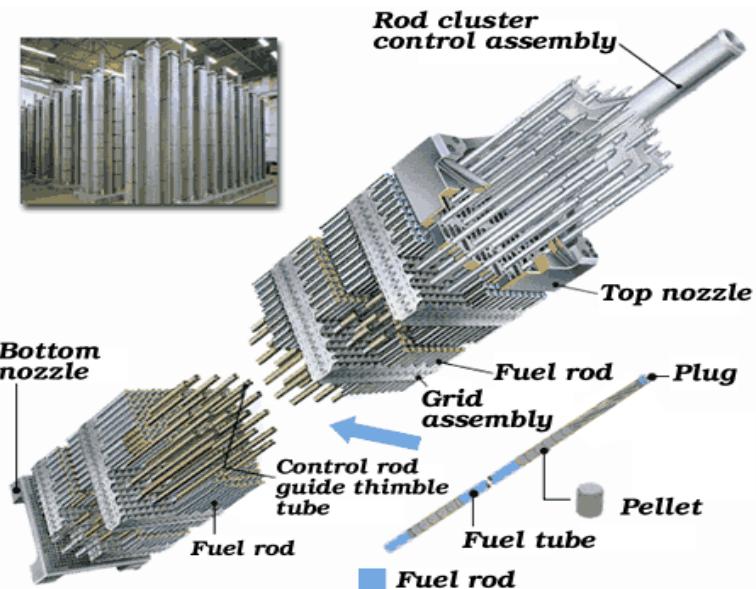
Physical Conditions

Temperatures: $400^{\circ} - 1200^{\circ}$ C
 $(\Delta T = 1000^{\circ}$ C / cm)

Damage Rate: $10^{-10} - 10^{-6}$ dpa/s

Stresses (approximate): ??

Fuel lifetime (in reactor): ~ 1 year

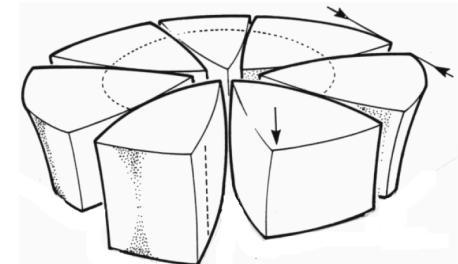
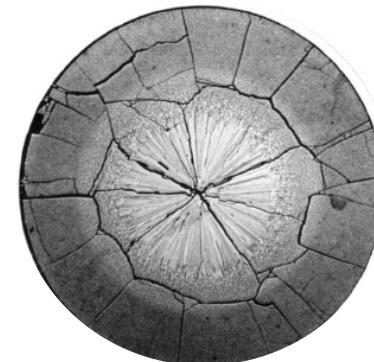


Fuel Swelling/Cracking

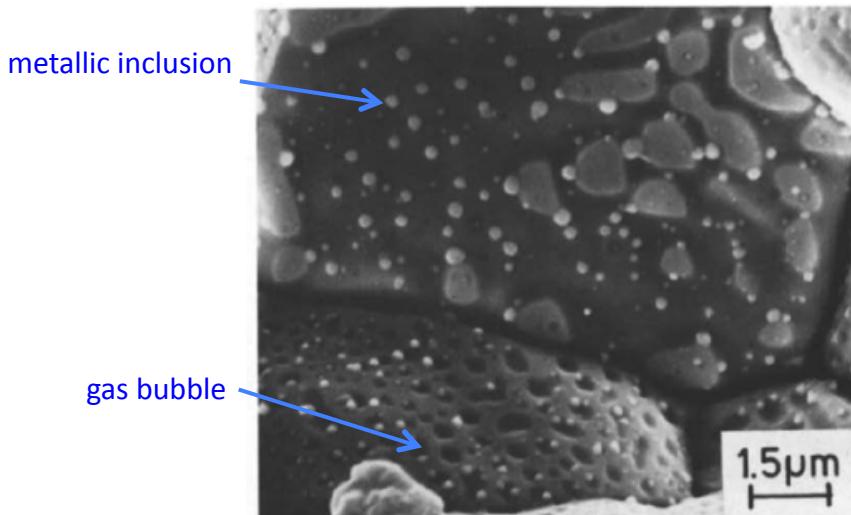
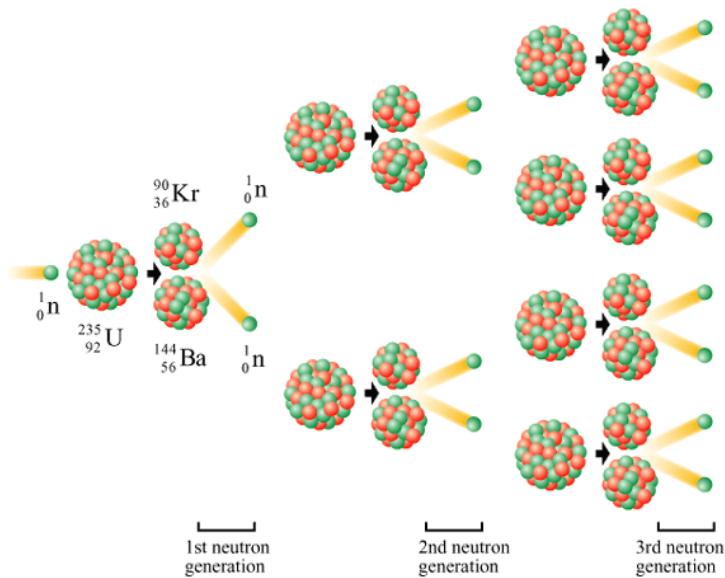
Non-uniform swelling due to temp. grad.

Radial cracks that form 'pie' slices

Occurs during initial power-up...



Fuel Microstructure



Walker et al., JNM 160 (1988) 10.

Fission Products

Solid: Ba, Zr, Mo, Cs, Sr, Ru, Nb,

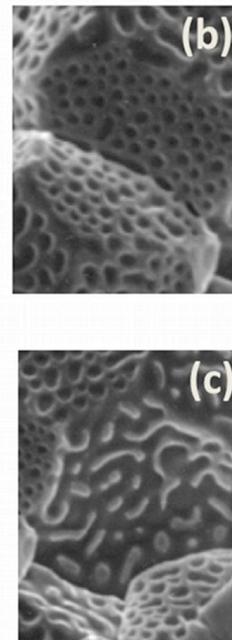
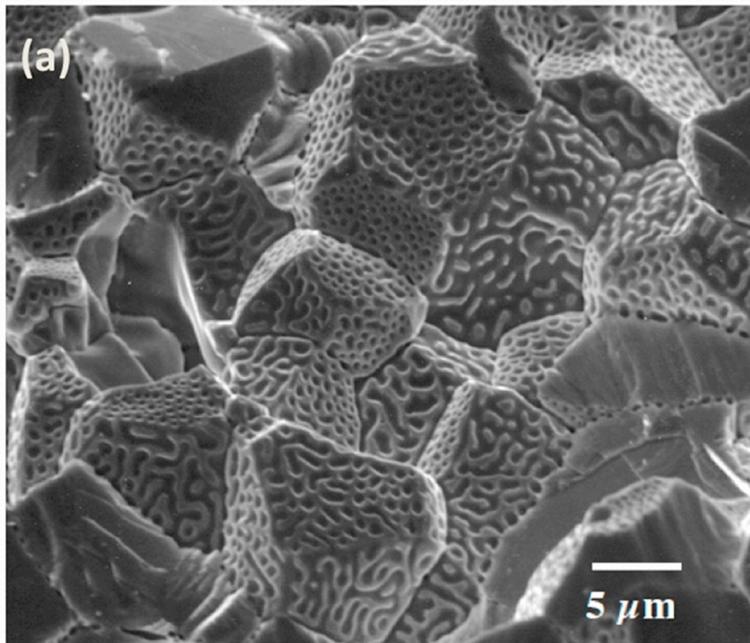
Gaseous: Kr, Xe

Importance...? fission products: (1) produce swelling, (2) change oxygen potential, (3) release gas {pressure on cladding}.

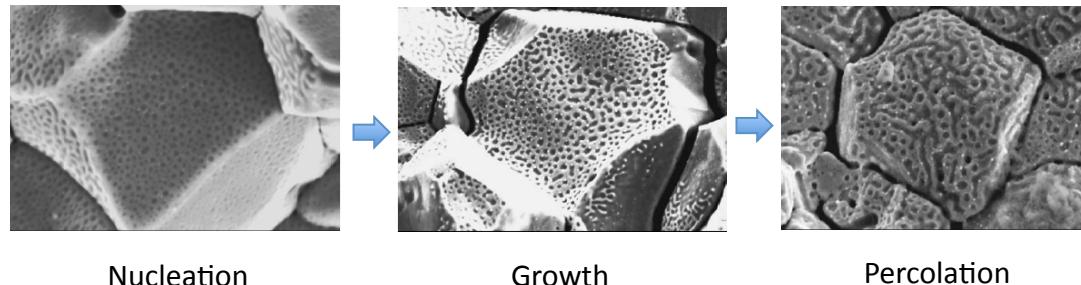
Precipitation

- Gas bubbles
- Metallic inclusions
- Oxide inclusions

Fuel Microstructure

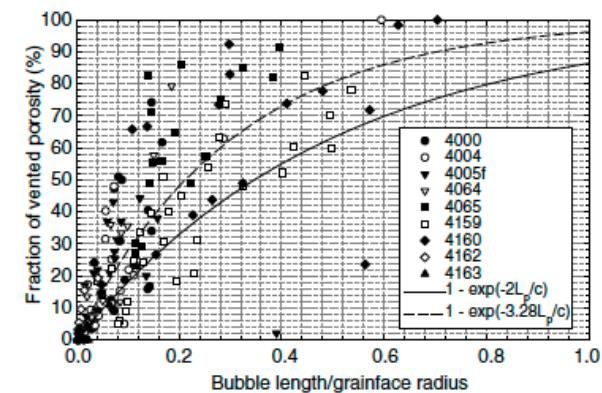


Valin et al. NEA Proc. 2000



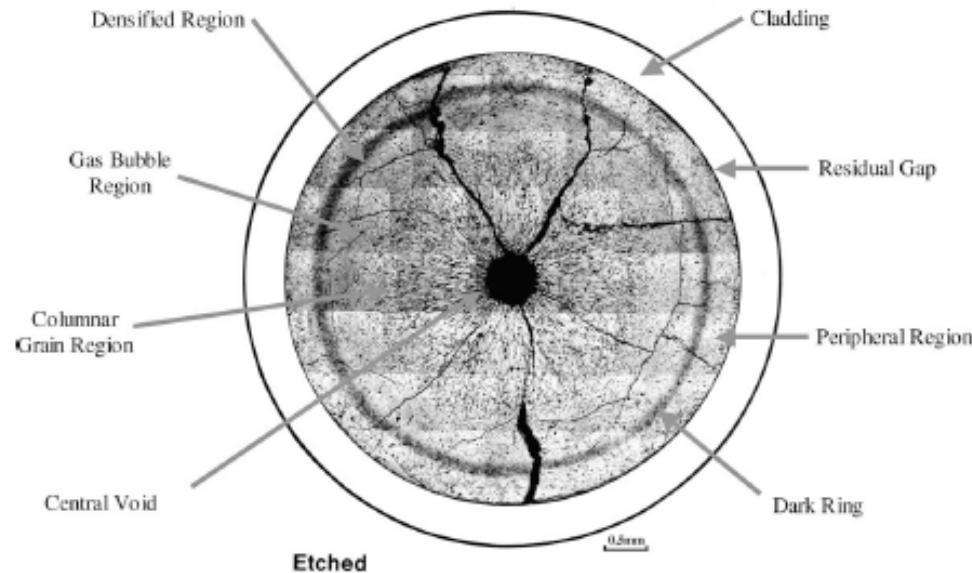
Intergranular Gas Bubbles

- Large bubble growth on grain boundaries
- Bubble percolation major mechanism of fission-gas release
- Bubble structure varies from GB to GB!



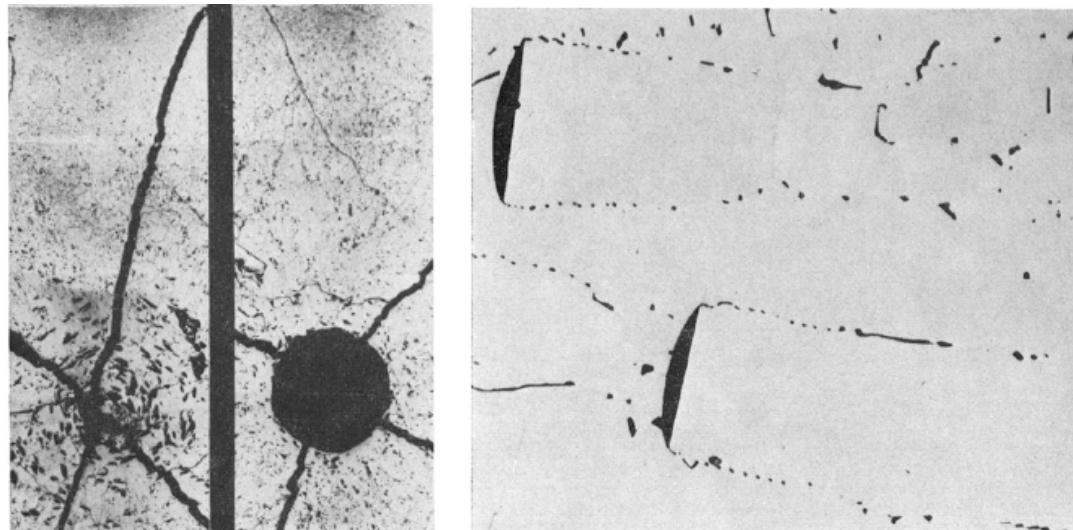
White, JNM 325 (2004) 61.

Fuel Microstructure



Fuel Restructuring in Thermal Gradient

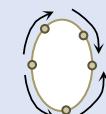
- Large temperature gradients drive microstructure evolution
 - Void migration in central region
 - Can create 'central void' & columnar grains
- (occurs in fast reactors, not PWRs)



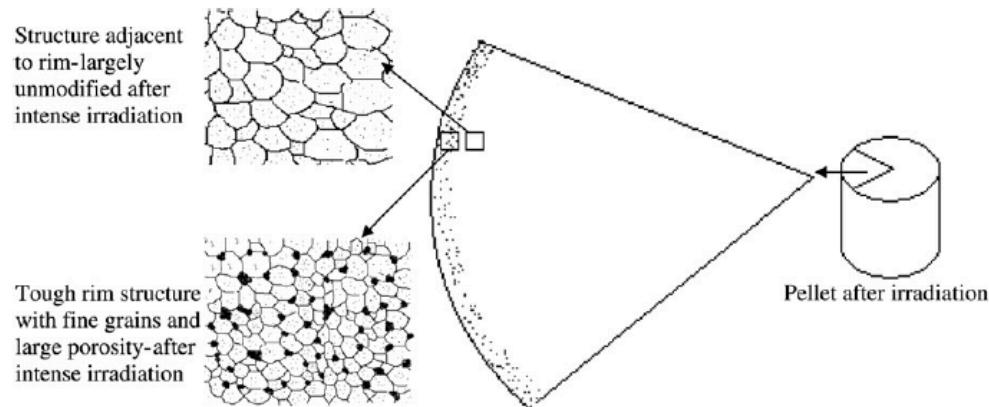
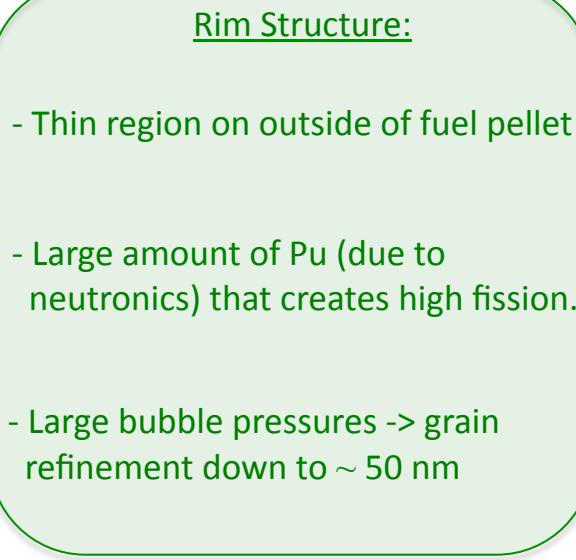
Sens, JNM 43 (1972) 293.

Mechanisms

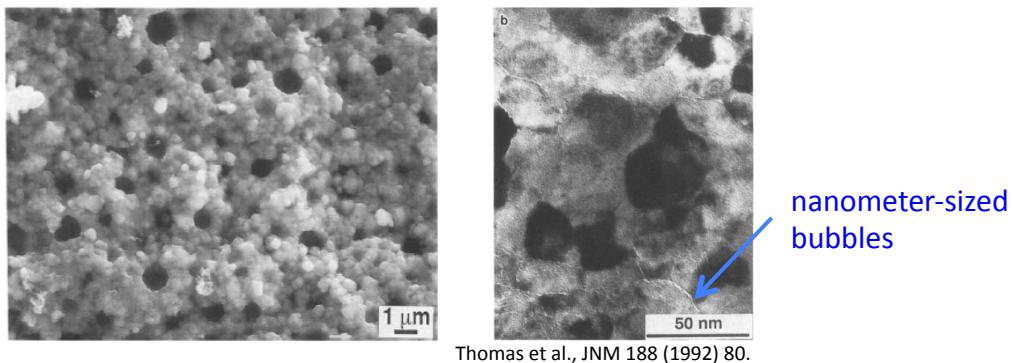
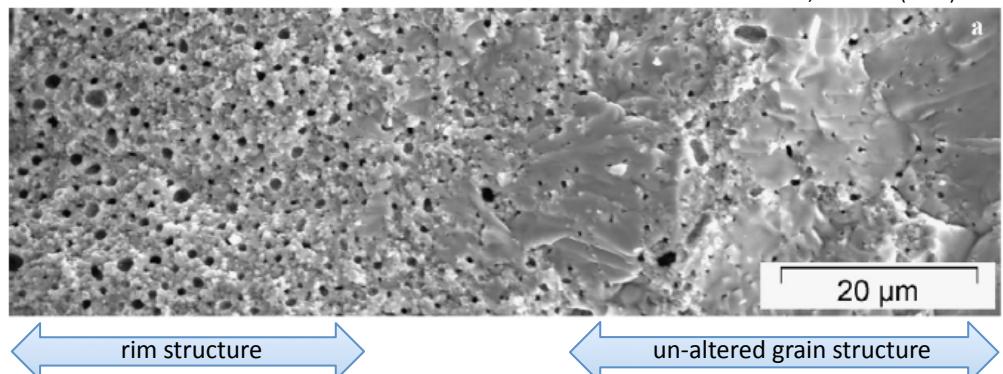
- Evaporation-condensation
- Thermo-migration



Fuel Microstructure



Noirot et al., JNM 372 (2008) 318.



Thomas et al., JNM 188 (1992) 80.

Meso-scale Modeling

Rate Theory

- Calculate time evolution of clusters
- Each cluster size gets own diff. eq.
- Takes into account defect production, cluster growth & shrinkage

•

$$\frac{\partial c_1}{\partial t} = P_1 - J_{1 \rightarrow 2} + J_{2 \rightarrow 1}$$

•

$$\frac{\partial c_2}{\partial t} = P_2 - J_{2 \rightarrow 3} + J_{3 \rightarrow 2} + J_{1 \rightarrow 2}$$

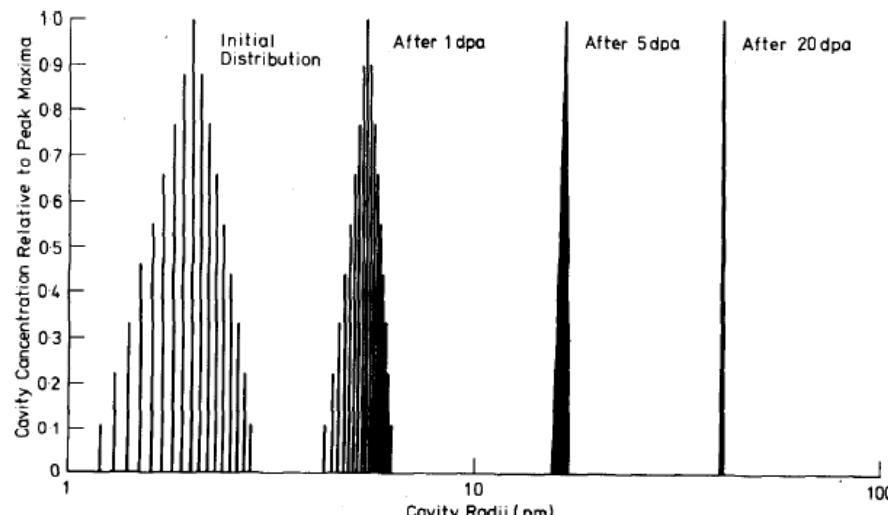
•

$$\frac{\partial c_3}{\partial t} = P_3 - J_{3 \rightarrow 4} + J_{4 \rightarrow 3} + J_{2 \rightarrow 3}$$

•

$$\frac{\partial c_4}{\partial t} = P_4 - J_{4 \rightarrow 5} + J_{5 \rightarrow 4} + J_{3 \rightarrow 4}$$

Can go up
to 1000's...



Hayns et al., JNM 87 (1979) 97.

Advantages:

- Concurrently capture nucleation and growth
- Straight-forward solution
- Multiple species & clusters (SIA clusters, gas)

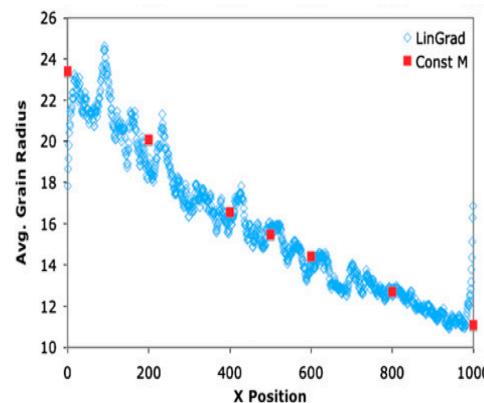
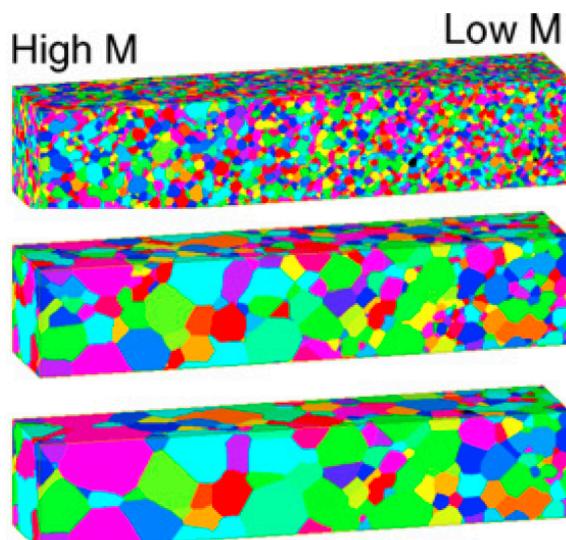
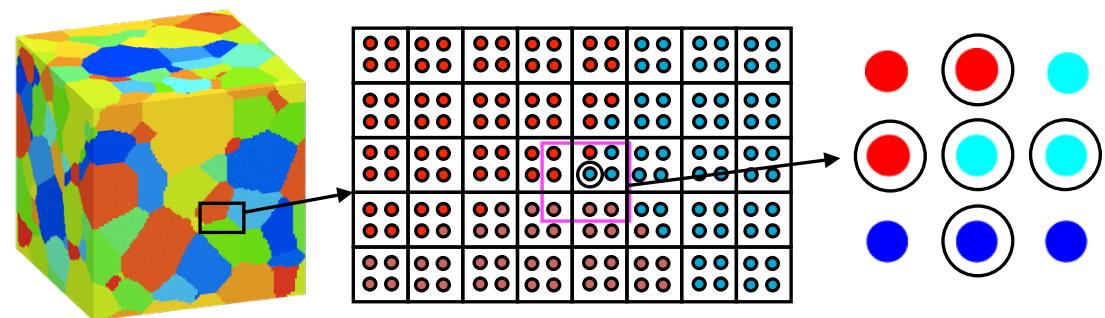
Disadvantages:

- Lacks spatial heterogeneity

Meso-scale Modeling

Potts Model

- Stochastic description of diffusion, interface motion
- Evolution driven by energy minimization
- Applied mainly to grain growth



Garcia et al., Scripta Mat. 59 (2008) 661.

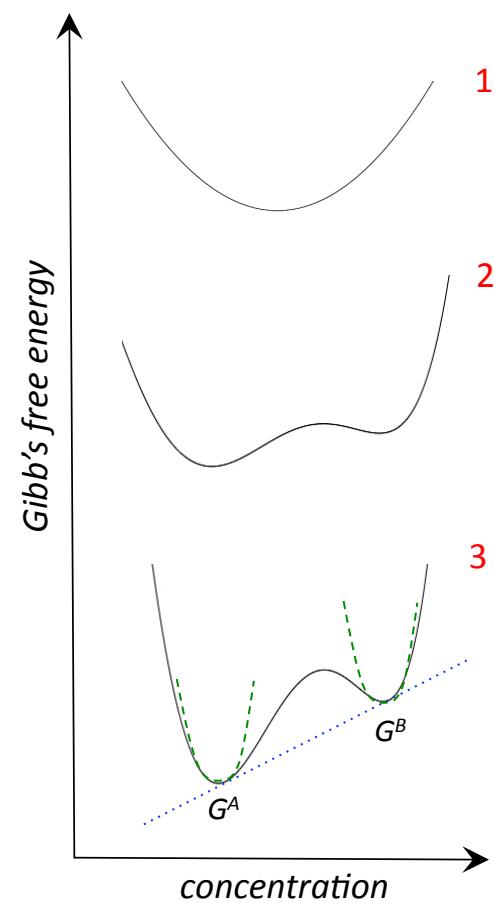
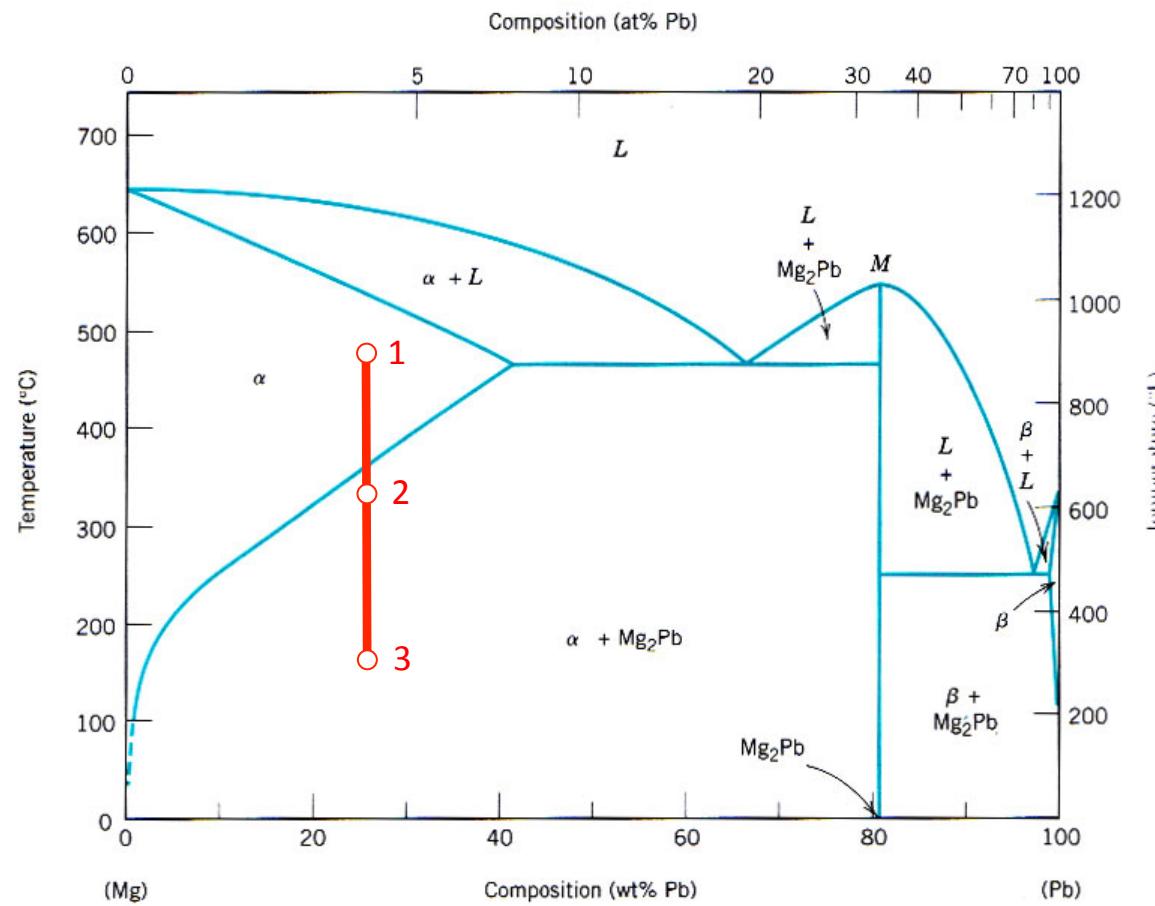
Advantages:

- Computationally efficient
- Applicable to variety of physics

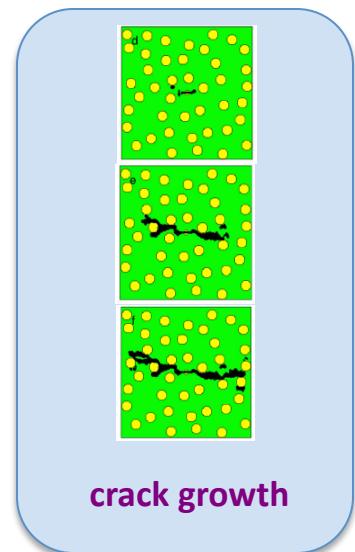
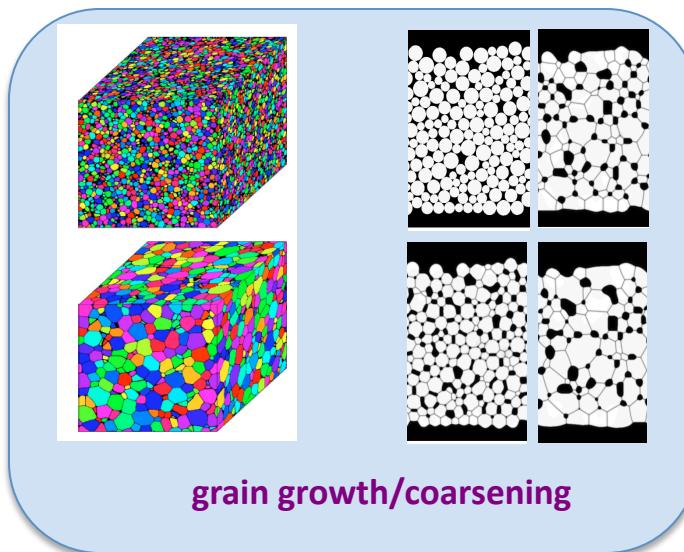
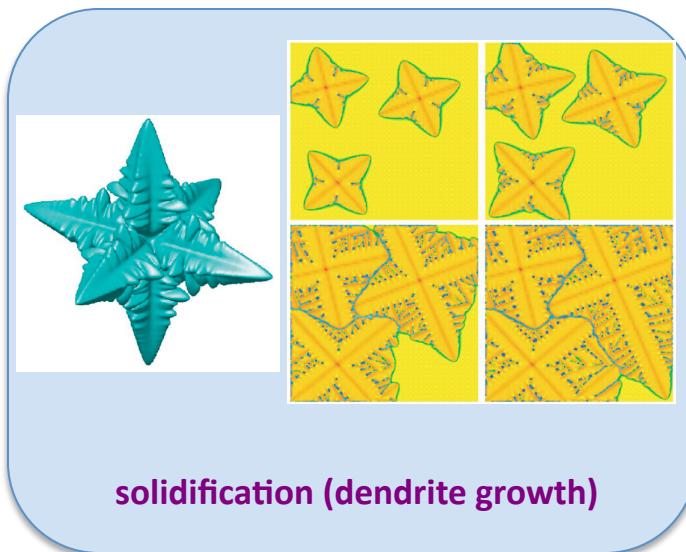
Disadvantages:

- Lacks gradients for diffusion
- Combining with FEM difficult...

Phase Transformations



Phase-Field Method



Species Diffusion:

conserved concentration fields:
 (vacancy, interstitial, solute concentrations)

$$\frac{\partial c_i(\mathbf{r},t)}{\partial t} = \nabla \cdot (M_{ij} \nabla \mu_i)$$

Cahn-Hilliard diffusion equation

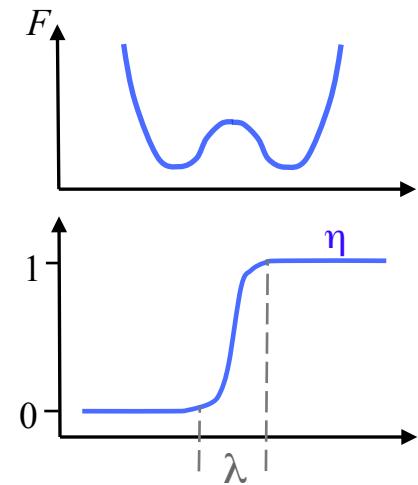
Nucleation and Interfacial Motion:

order parameters that represent:
 (precipitates, voids, gas bubbles, grains)

$$\frac{\partial \eta_i(\mathbf{r},t)}{\partial t} = -L_i \frac{\delta F}{\delta \eta_i(\mathbf{r},t)}$$

Allen-Cahn equation

Diffuse interface:



Governing equations:

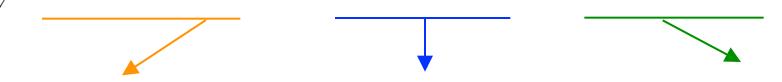
$$\left\{
 \begin{array}{l}
 \frac{\partial c_v}{\partial t} = \nabla \cdot \left(M_v \nabla \frac{1}{N} \frac{\delta F}{\delta c_v} \right) + \xi + P_v - R_{iv} - S_v^{GB} \quad (\text{vacancies}) \\
 \frac{\partial c_i}{\partial t} = \nabla \cdot \left(M_i \nabla \frac{1}{N} \frac{\delta F}{\delta c_i} \right) + \zeta + P_i - R_{iv} - S_i^{GB} \quad (\text{interstitials}) \\
 \frac{\partial c_g}{\partial t} = \nabla \cdot \left(M_g \nabla \frac{1}{N} \frac{\delta F}{\delta c_g} \right) + \gamma + P_g \quad (\text{gas atoms})
 \end{array}
 \right.$$

CH

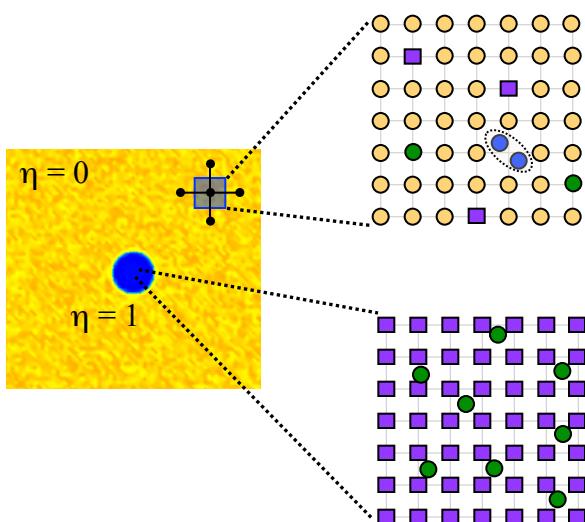
AC

Free energy

$$F = N \int_V [h(\eta) f^{\text{solid}}(c_v, c_i, c_g) + j(\eta) f^{\text{bubble}}(c_v, c_i, c_g) + f^{\text{int}}(c_v, c_i, c_g, \eta)] dV$$



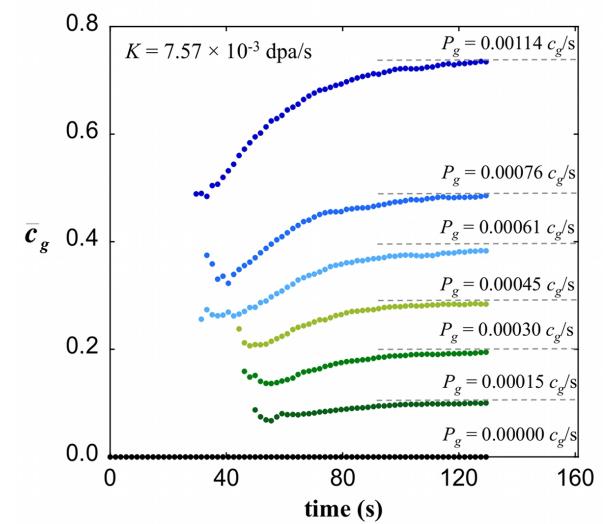
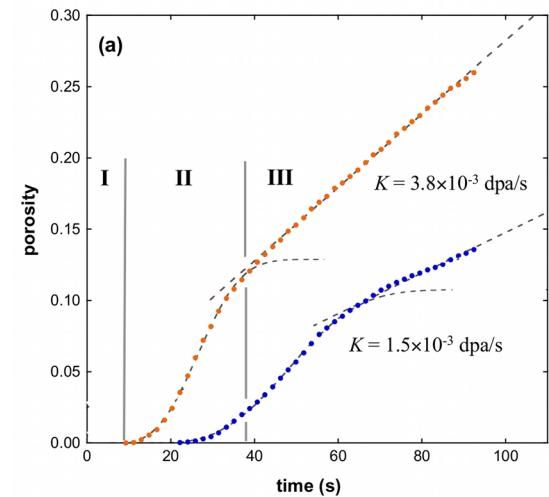
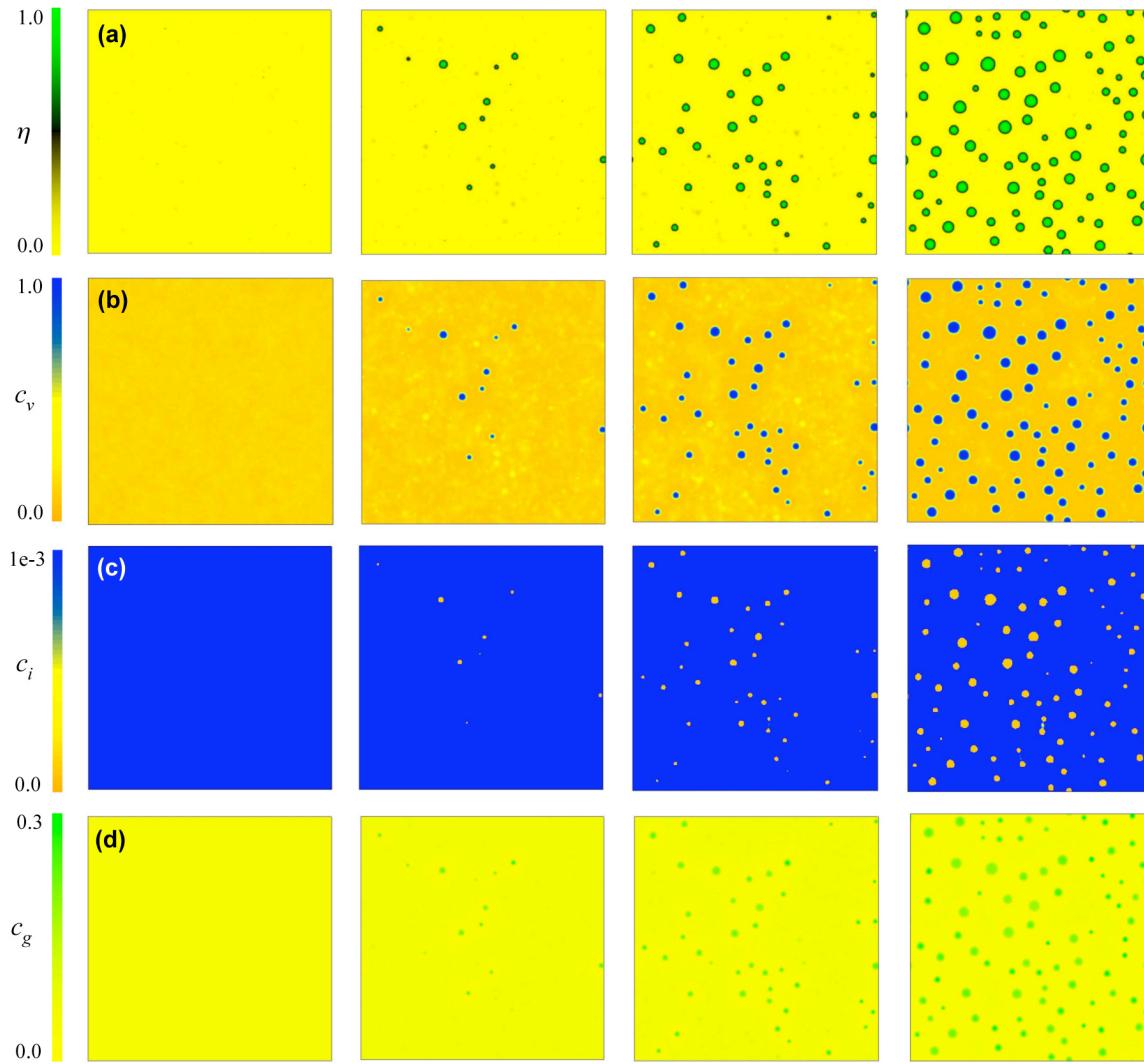
 solid bubble interfacial



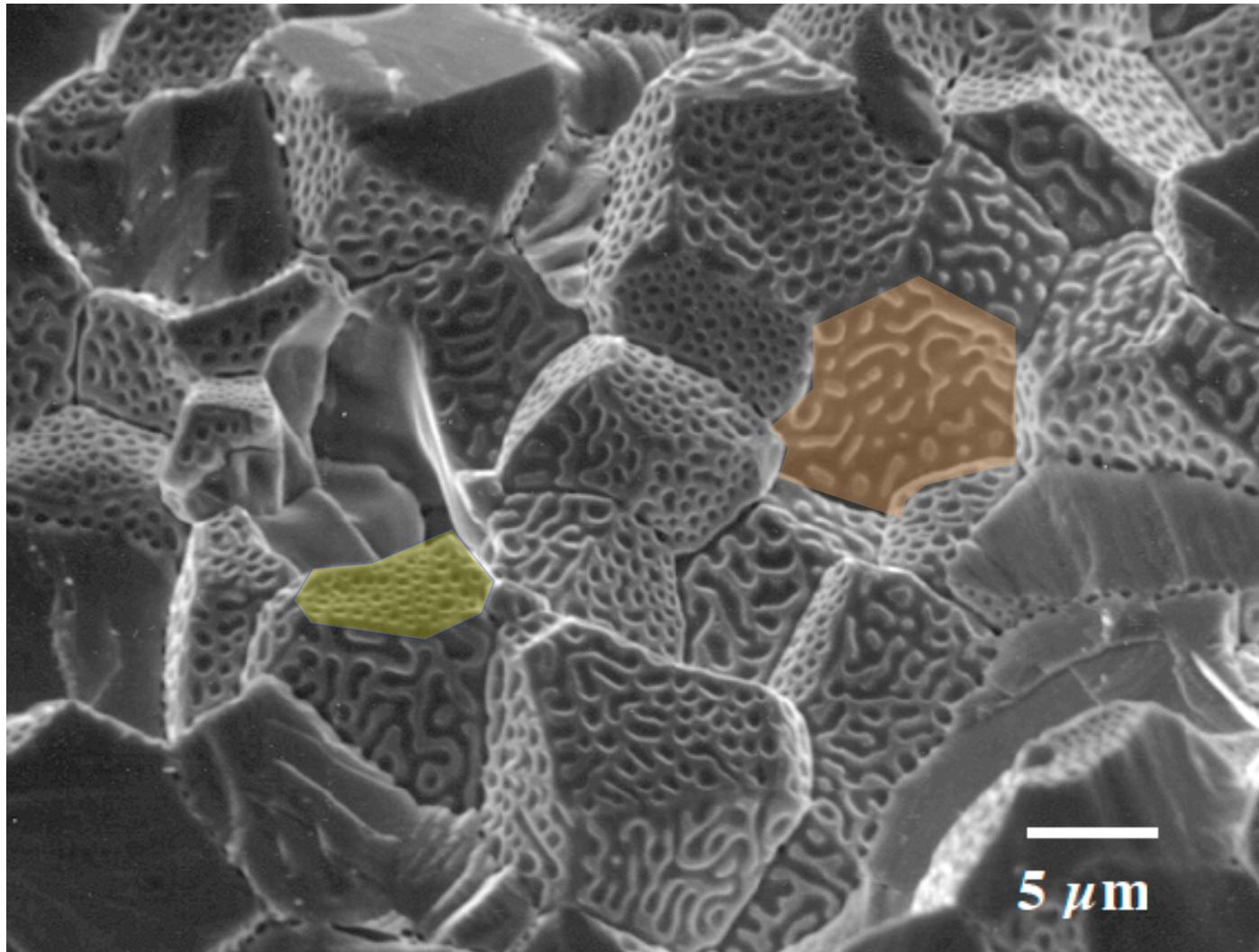
$$f^{\text{solid}} = \underbrace{E_v^f c_v + E_i^f c_i + E_g^f c_g}_{\text{enthalpy}} + \underbrace{k_B T [c_v \ln(c_v) + c_i \ln(c_i) + c_g \ln(c_g) + (1 - c_v - c_i - c_g) \ln(1 - c_v - c_i - c_g)]}_{\text{configurational entropy}}$$

$$f^{\text{bubble}}(c_v, c_i, c_g) = \underbrace{[(c_v - 1)^2 + c_i^2]}_{\text{Landau term}} + \underbrace{[\mu_g^o c_g + c_g k_B T \ln c_g + c_g k_B T \ln(k_B T)]}_{\text{ideal gas law}}$$

Bubble Kinetics

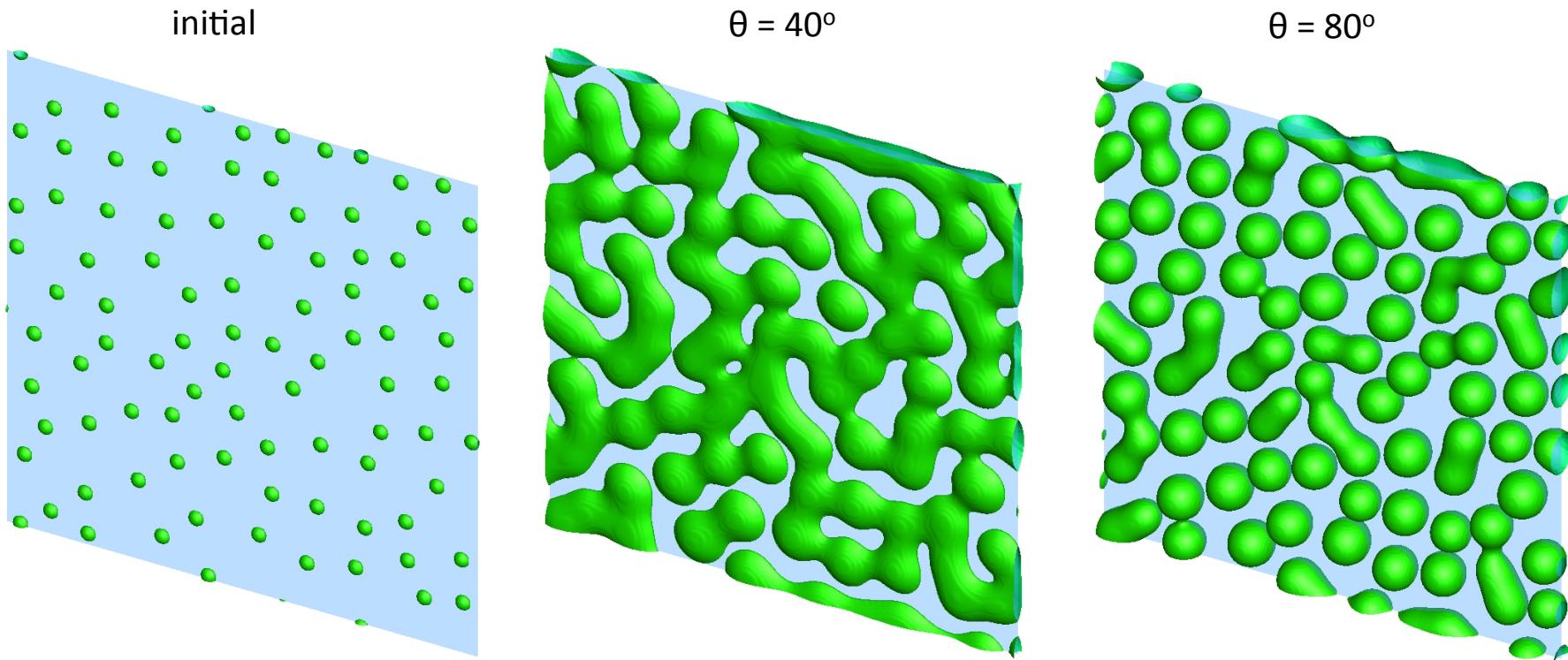


Fission Gas Release



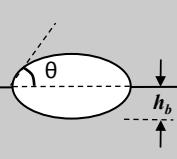
Why do some grain boundaries have percolated bubble structures, while others do not?

Fission Gas Release

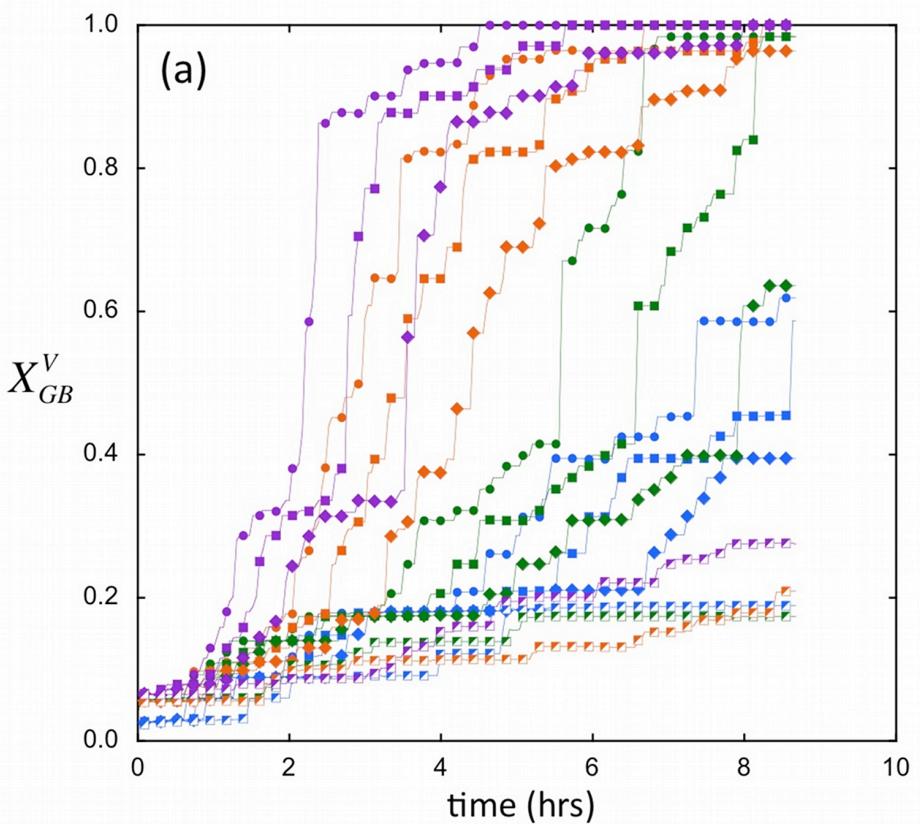
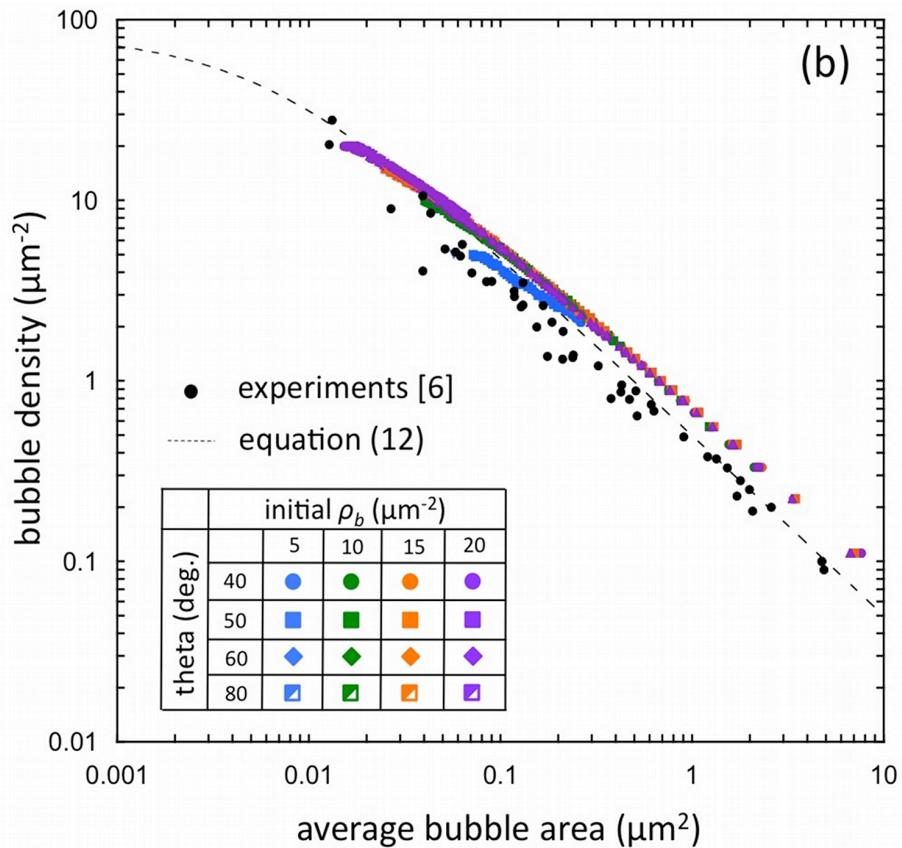


Phase-field Simulation of GB Bubble Growth:

- Bicrystal with initial number density of bubbles on grain boundary.
- Vary contact angle (θ), nucleation density, & GB gas diffusivity.



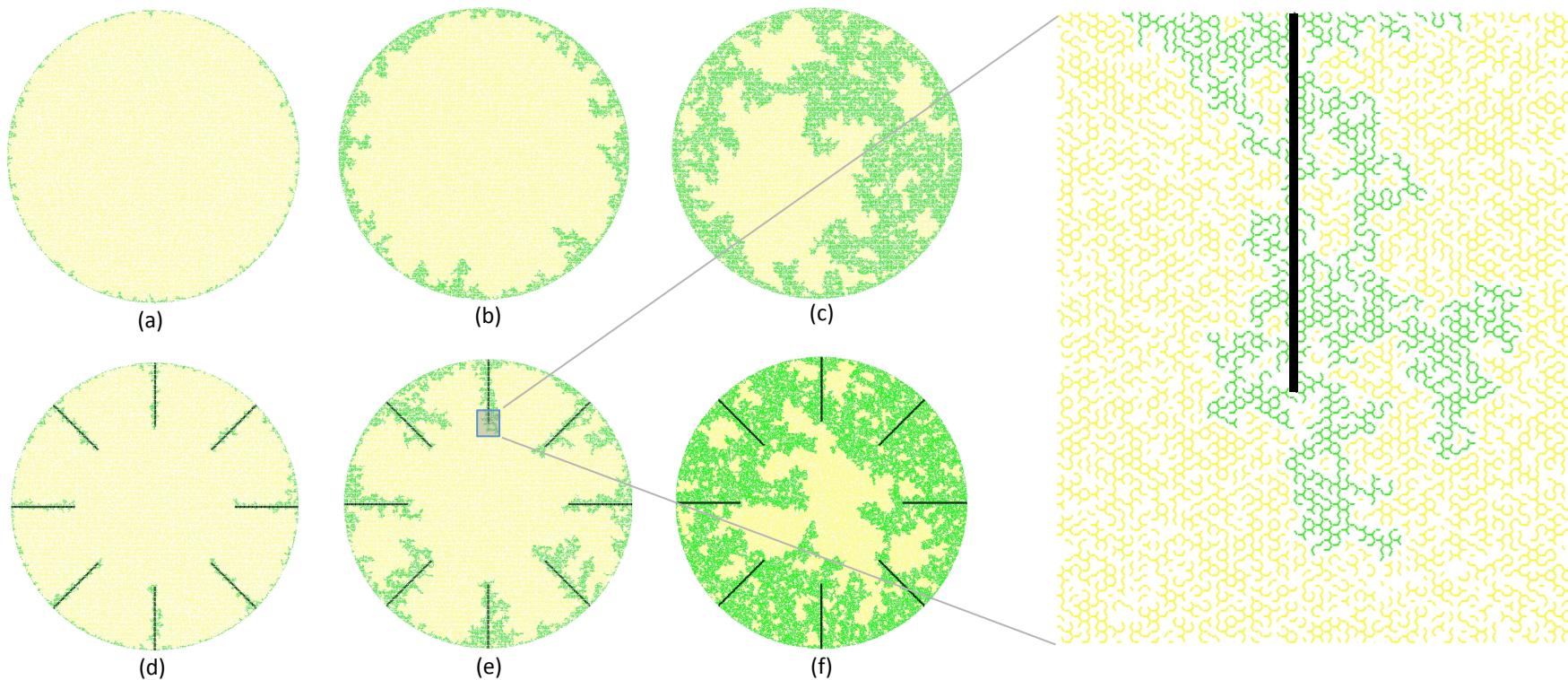
Fission Gas Release



Good agreement with experiments

Large discrepancy in percolation rates!

Fission Gas Release

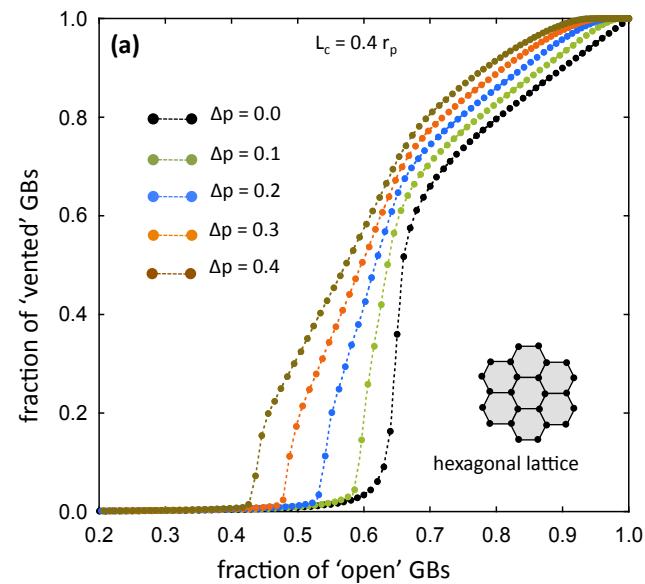
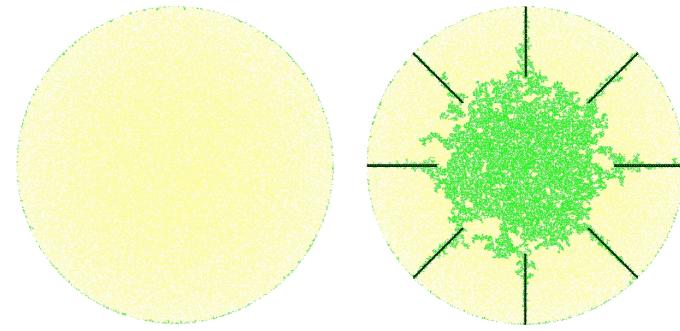
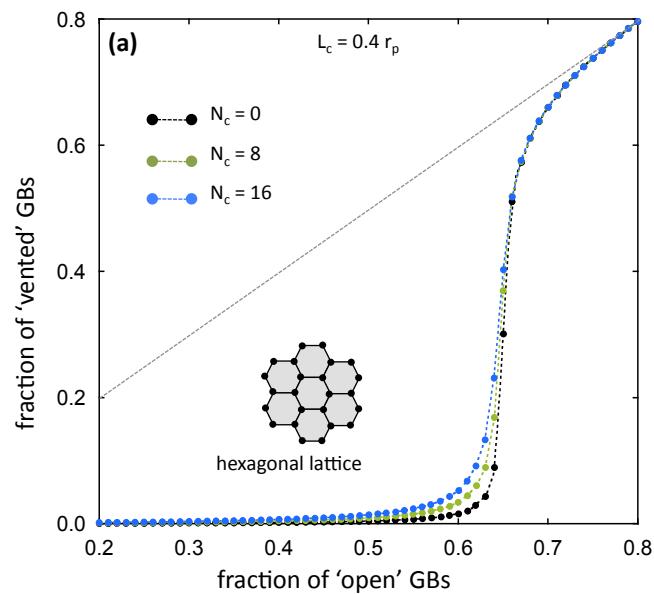
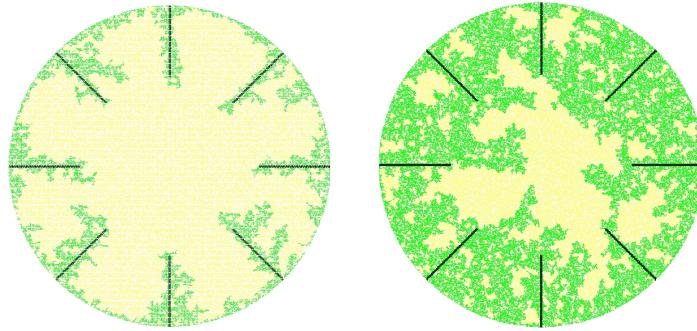


Percolation Modeling:

- Assume a network of GBs
- Randomly assign status of 'open' or 'closed'
- Search for 'open' GBs that connect to free surface

Can also include cracks!

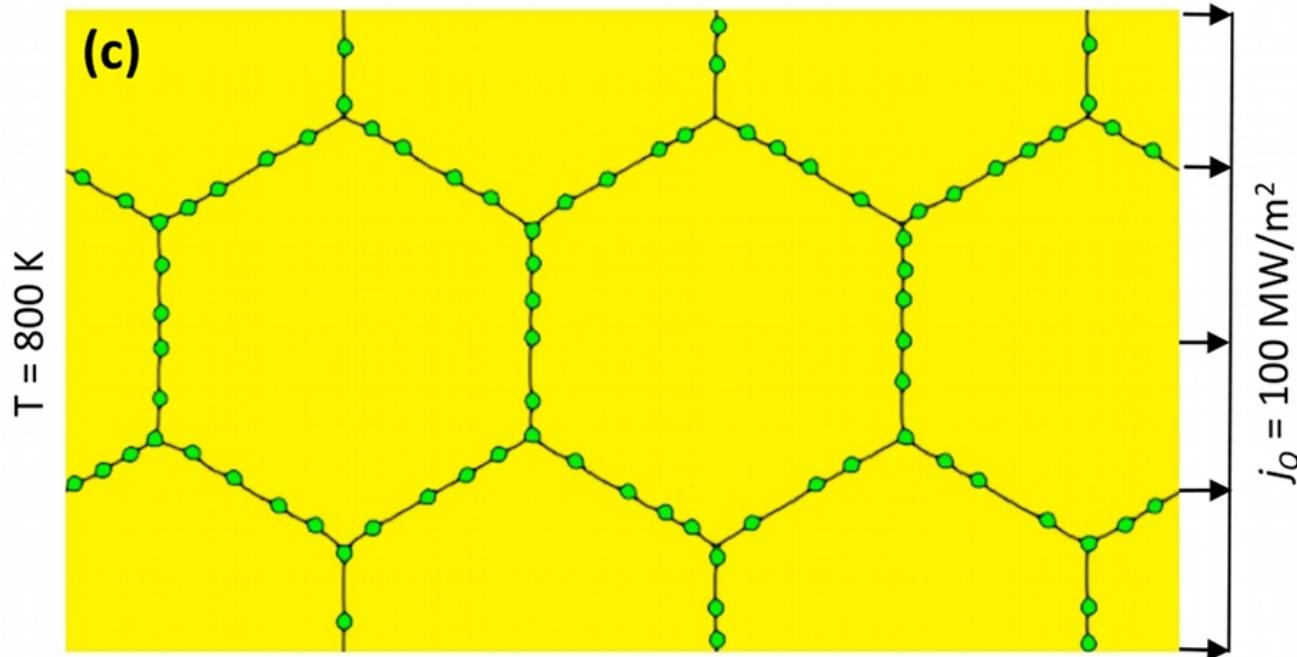
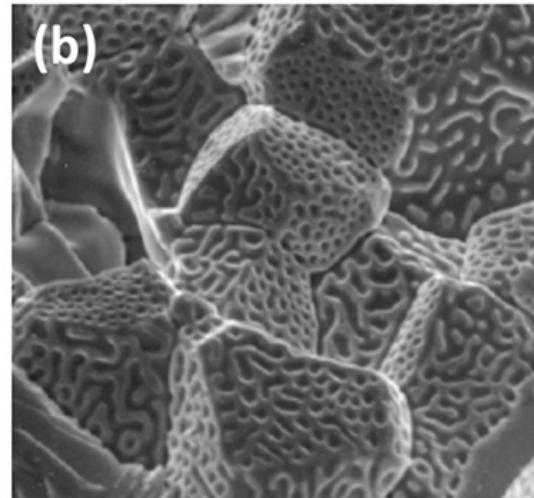
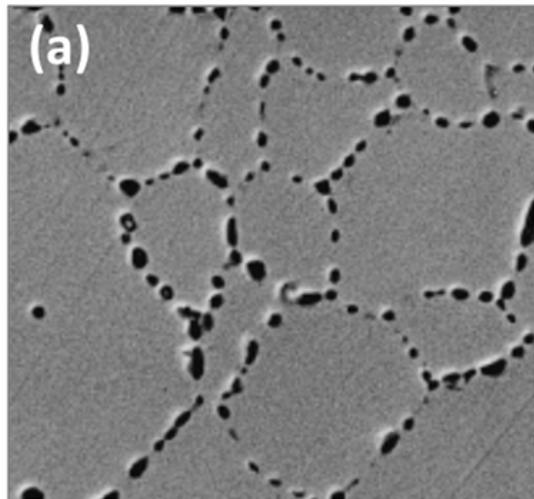
Fission Gas Release



Percolation threshold ~ 0.64
Cracks make little difference!

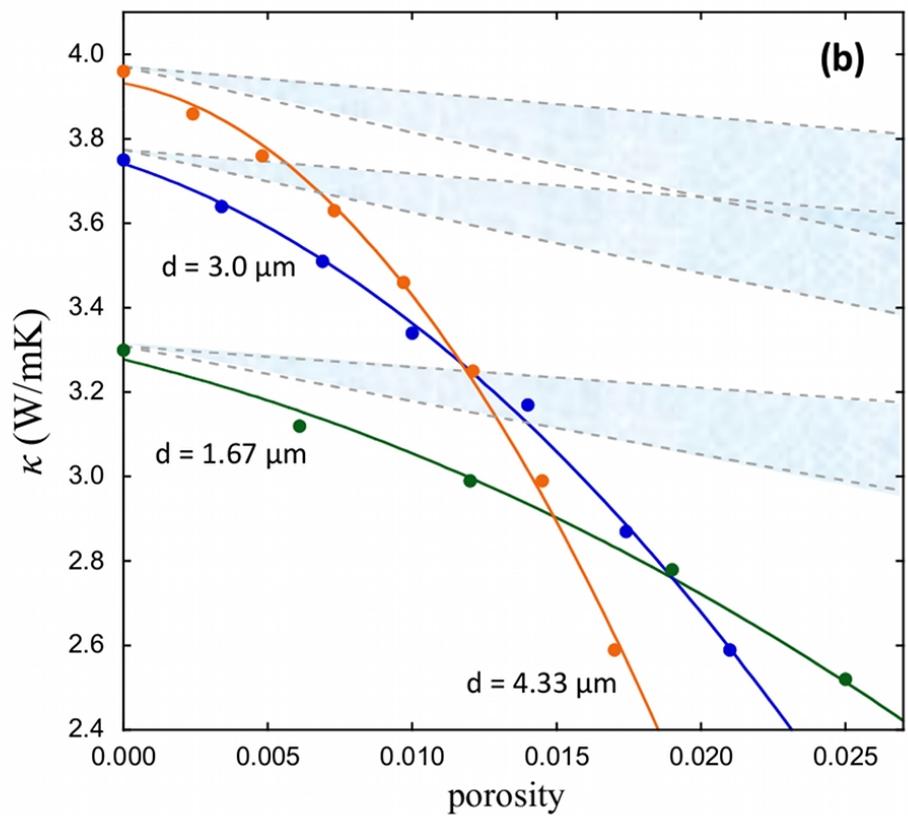
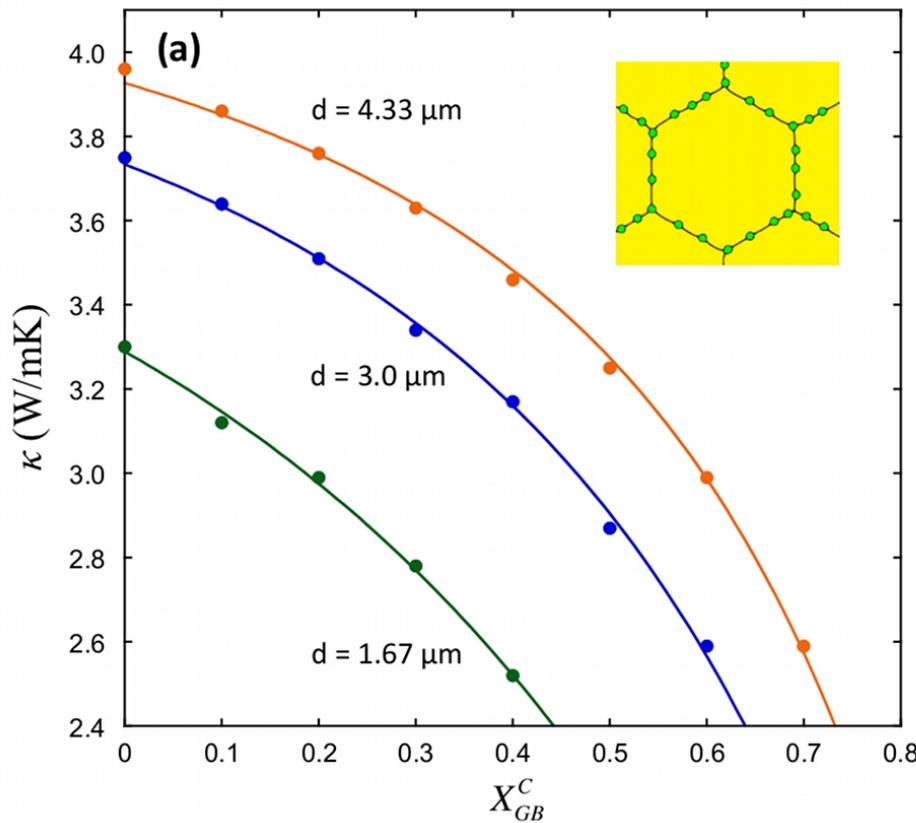
...But, with a gradient in percolation fraction, cracks do matter!

Thermal Conductivity



Goal: To determine the effect of GB bubbles on thermal conductivity

Thermal Conductivity



Conductivity versus
GB coverage

Large discrepancy with traditional
models!